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REPORT DOCUMENTATION PAGE

Form Approved OMB NO. 0704-0188

1. AGENCY USE ONLY *(Leave blank)*

2. REPORT DATE
May 15, 1998

3. REPORT TYPE AND DATES COVERED
Final Technical Report

4. TITLE AND SUBTITLE
Correlation Effects in Multiple Scattering from Random Rough Surfaces

5. FUNDING NUMBERS
DAAH04-94-G-0325

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8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES)

U.S. Army Research Office
P.O. Box 12211
Research Triangle Park, NC 27709-2211

10. SPONSORING / MONITORING AGENCY REPORT NUMBER
ARO 32304.12-PH

11. SUPPLEMENTARY NOTES
The views, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy or decision, unless so designated by other documentation.

12a. DISTRIBUTION / AVAILABILITY STATEMENT

Approved for public release; distribution unlimited.

19981222 074

13. ABSTRACT *(Maximum 200 words)*
The first theoretical studies of angular correlations in the intensity of light scattered from randomly rough metal surfaces, that go beyond the factorization approximation, have been carried out. As a consequence, in addition to the memory and the time-reversed memory effects studied by earlier authors, known as the short-range $C^{(1)}$ correlations, the long-range $C^{(2)}$ correlations and the infinite-range $C^{(3)}$ correlations have been determined. The latter two correlation functions had been studied earlier in the scattering of light from volume disordered systems, but had not yet been studied in rough surface scattering. In addition, a new correlation function, called $C^{(10)}$, of the same order of magnitude as $C^{(1)}$ has been found, as well as another new correlation function, called $C^{(1.5)}$, that, along with $C^{(2)}$, displays peaks associated with the roughness-induced excitation of the surface electromagnetic waves supported by the vacuum-metal interface. The invalidity of the factorization approximation for weakly rough random surfaces has also been established in another set of calculations. Experimental studies of the memory and time-reversed memory effects have been carried out, as have studies of the effects of localized defects on angular correlations in the intensity of light scattered from random surfaces.

14. SUBJECT TERMS
angular intensity correlation functions, surface roughness, enhanced backscattering, satellite peaks, metal films, dielectric films, memory effect, time-reversed memory effect

15. NUMBER IF PAGES
15

16. PRICE CODE

17. SECURITY CLASSIFICATION OR REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
UNCLASSIFIED	UNCLASSIFIED	UNCLASSIFIED	UL

Summary of the most important results

During the period of the grant 18 papers were published or submitted for publication. We summarize here the most important results obtained.

A. Giant Enhanced Backscattering of Light

The enhanced backscattering of light from a randomly rough surface, which is manifested as a narrow peak in the retroreflection direction in the angular distribution of the intensity of the light that is scattered diffusely, has been extensively studied. Both theoretical and experimental investigations have shown that the height of the peak is never more than twice the height of the background at the position of the peak. In a recent paper(1)* we reported the observation of a giant enhanced backscattering of light from a randomly weak rough dielectric film on a reflecting metal substrate, in which the ratio of the height of the peak to the height of the background at its position is greater than 10. It was found that this giant enhanced backscattering peak is accompanied by concentric circular interference fringes, whose axis is normal to the mean scattering surface,with both the specular and backscattering peak on the same ring. A possible mechanism for the giant enhanced backscattering was suggested.

The design of laser radars and the determination of laser radar signatures of various vehicles require accurate data on the reflectivity of the target materials to far-field laser illumination at many different wavelengths. Modelling software is required that will take the monostatic reflectance data as input, and combine this data with geometrical models to develop a laser radar signature. Consideration of enhanced backscattering can be critically important in developing laser radar signatures. This is illustrated in a second paper(2) by the comparison of two signature models of the same airplane– one developed using bidirectional reflectance measurement data at two degrees from the retro-direction, and the other developed from monostatic measurement data at true retro-direction. When a reflecting metal substrate is coated with a thin layer of a weakly rough dielectric, as is often the case with naturally occurring metal oxides on a parent metal surface, a giant enhanced backscattering peak is observed which further enhances the ladar signature.

In a third paper(3) an optical method of fabricating one- and two-dimensional diffrac-

*The references are to papers included in the "List of all publications" below.

tion gratings is described. As opposed to conventional gratings which feature regularly spaced grooves, these gratings have variable profiles which represent mathematical functions previously defined. These surface profiles are produced by a laser beam exposing photoresist-coated plates with a raster scanner under computer control. Two kinds of 1-D profiles for the gratings have been made: deterministic–the profile represents a mathematical function; random-gratings possess a random profile with certain statistical distributions. Several surfaces have been fabricated and characterized with a stylus profilometer. By changing the deterministic and statistical properties of fabricated surfaces, the light-scattering in the far field can be controlled for many optical applications.

B. Theoretical Studies of Angular Intensity Correlation Functions for Light Scattered from Weakly Rough Random Metal Surfaces

In the majority of the theoretical and experimental studies of phenomena occurring in the multiple scattering of electromagnetic waves from randomly rough surfaces, the quantity that has been studied is an intensity, a second moment of the reflected or transmitted field. Recently, however, attention has begun to be directed toward theoretical and experimental studies of higher-order moments of these fields, in particular of angular intensity correlation functions. This interest has been stimulated by the expectation that, just as the inclusion of multiple-scattering processes into the calculation of the angular dependence of the intensity of the incoherent component of the field scattered from a randomly rough surface led to the prediction of enhanced backscattering, their inclusion in the calculation of higher moments of the scattered field will also lead to the prediction of new physical effects.

This expectation was prompted by the results of theoretical and experimental investigations of intensity correlation functions in the scattering of classical waves from volume disordered media. In theoretical investigations it was predicted that three types of intensity correlations occur, namely short-range correlations, long-range correlations, and infinite-range correlations. These have been termed the $C^{(1)}$, $C^{(2)}$, and $C^{(3)}$ correlations, respectively. The $C^{(1)}$ correlation function includes both the "memory effect" and the "time-reversed memory effect," so named because of the wave vector conservation conditions they satisfy. Both of these effects have now been observed in volume scattering experiments. The $C^{(2)}$

correlation function has also been observed in such experiments, while the $C^{(3)}$ correlation function has been seen only in experiments on volume disordered media in the microwave frequency range.

Until recently, only the $C^{(1)}$ correlation function, namely the memory and time-reversed memory effects had been studied theoretically and experimentally in the scattering of light from randomly rough surfaces. However, in a recent series of papers devoted to studies of angular correlation functions of the intensity of light scattered from one-dimensional[4–6] and two-dimensional[5] randomly rough metal surfaces the $C^{(2)}$ and $C^{(3)}$ correlation functions have been calculated, and two additional types of correlation functions named the $C^{(10)}$ and $C^{(1.5)}$ correlation functions have been predicted.

The angular intensity correlation function studied in these papers is defined by

$$\begin{aligned} C(q,k|q',k') &= \langle [I(q|k) - \langle I(q|k)\rangle][I(q'|k') - \langle I(q|k')\rangle]\rangle \\ &= \langle I(q|k)I(q'|k')\rangle - \langle I(q|k)\rangle\langle I(q'|k')\rangle \end{aligned} \tag{1}$$

for p-polarized light of frequency ω scattered from a weakly rough one-dimensional random metal surface. $I(q|k)$ is given by $I(q|k) = (\omega/c)|S(q|k)|^2/L$, where $S(q|k)$ is the scattering matrix for the scattering of light the projection of whose wave vector on the mean scattering surface is k into light the projection of whose wave vector on the mean scattering surface is q, and L is the length of the x_1-axis covered by the random surface. The angle brackets denote an average over the ensemble of realizations of the surface profile function.

Since the correlation of an intensity with itself is stronger than the correlation of two different intensities a peak in $C(q,k|q',k')$ is expected when $q = q'$ and $k = k'$. This is the memory effect. Because $S(q|k)$ is reciprocal, $S(q|k) = S(-k|-q)$, a peak in $C(q,k|q',k')$ is also expected when $q = -k'$ and $k = -q'$. This is the time-reversed memory effect.

In terms of $S(q|k)$ the correlation function $C(q,k|q',k')$ becomes

$$\begin{aligned} C(q,k|q',k') &= \frac{1}{L^2}\frac{\omega^2}{c^2}[\langle S(q|k)S^*(q|k)S(q'|k')S^*q'|k')\rangle - \\ &\quad -\langle S(q|k)S^*(q|k)\rangle\langle S(q'|k')S^*(q'|k')\rangle]. \end{aligned} \tag{2}$$

A commonly used approximation to this expression is based on the *factorization approximation*, which is obtained by writing $C(q,k|q',k')$ as

$$C(q,k|q',k') \simeq \frac{1}{L^2}\frac{\omega^2}{c^2}\left[|\langle S(q|k)S^*(q'|k')\rangle|^2 + |\langle S(q|k)S(q'|k')\rangle|^2\right], \tag{3}$$

and neglecting the second term. Both of these steps are valid if the second-order statistics of the scattered field is Gaussian and circular, and the resulting approximation

$$C(q,k|q',k') \simeq \frac{1}{L^2}\frac{\omega^2}{c^2}|\langle S(q|k)S^*(q'|k')\rangle|^2 \tag{4}$$

is sufficient for predicting the memory and time-reversed memory effects.

In the work described in Refs. 4 and 5 the factorization approximation was not made. In effect, the scattering matrix $S(q|k)$ was expanded in powers of the surface profile function $\zeta(x_1)$, assumed to be a zero-mean, stationary Gaussian random process, the right-hand side of Eq. (2) was then expanded in the same way, and the averaging was carried out term-by-term. The lowest order contributions are of $O(\zeta^4)$; there are two of them, one proportional to $\delta(q-k-q'+k')$, the other to $\delta(q-k+q'-k')$. They are multiplied by structureless functions of the four wave numbers. They are in fact the lowest order contributions to the two terms on the right-hand side of Eq. (3). In sixth order terms proportional to the same two delta-functions also occur (indeed such terms occur in every nonzero order in $\zeta(x_1)$). The term proportional to $\delta(q-k-q'+k')$ is multipled by an envelope function that contains peaks at $q=q'$ and $k=k'$ and at $q=-k'$ and $k=-q'$. The term proportional to $\delta(q-k+q'-k')$ is multiplied by a smoothly varying function of the four wave numbers. The terms proportional to $\delta(q-k-q'+k')$ constitute the $C^{(1)}$ correlation function; the terms proportional to $\delta(q-k+q'-k')$ constitute a new correlation function, denoted by $C^{(10)}$, which is not captured by the factorization approximation. In eighth order the lowest order contribution to the correlation function $C^{(2)}$ is obtained. There are no restrictions on the four wave numbers in this contribution. It displays peaks when conditions such as $q'=-k$ (without the condition $q=-k'$ having also to be satisfied, as in $C^{(1)}$), $q'=q$, $q'=-q$, and $q'=k$, are satisfied. These peaks are associated with the roughness-induced excitation of the surface plasmon polaritons supported by the vacuum-metal interface. The leading contribution to $C^{(3)}$ arises in the tenth order, but the twelfth-order contribution to it dominates. $C^{(3)}$ is a

smoothly varying function of q, k, q', k' with no structure to it. The assignment of the various contributions to $C(q, k|q', k')$ to the $C^{(1)}, C^{(2)}$ and $C^{(3)}$ correlation functions is based on the degree to which the scattered waves retain a "memory" of the waves incident on the random surface. In sixth order a second new correlation function first makes its appearance, that is termed the $C^{(1.5)}$ correlation function, because its lowest order contribution is intermediate between the lowest order contributions to $C^{(1)}$ and $C^{(2)}$. $C^{(1.5)}$, which also is not captured by the factorization approximation, has no restrictions on the wave numbers q, h, q', k'. However, it displays peaks when conditions such as $-k' + k + q' = -k_{sp}(\omega)$ and $q - q' + k' = k_{sp}(\omega)$ are satisfied, where $k_{sp}(\omega)$ is the wave number of the surface plasmon polariton at the vacuum-metal interface at the frequency of the incident light, and are associated with the excitation of surface plasmon polaritons.

Although the preceding discussion has been presented in the context of the scattering of light from a one-dimensional, randomly rough, metal surface, the angular intensity correlation functions calculated for the scattering of light from two-dimensional random metal surfaces also display the $C^{(1)}, C^{(10)}, C^{(1.5)}, C^{(2)}$, and $C^{(3)}$ correlations[5].

Thus, the angular intensity correlations of light scattered from one- and two-dimensional, randomly rough, metal surfaces display a rich structure that is estimated to be observable experimentally. It is hoped that the theoretical work presented in Refs. 4-6 will stimulate efforts to observe this structure. This work also demonstrates that the use of the factorization approximation in calculations of these correlation function results in interesting features being overlooked, namely the $C^{(10)}$ and $C^{(1.5)}$ correlations.

C. Experimental Studies of Angular Intensity Correlation Functions

The memory effect and the time-reversed memory effect in the angular intensity correlation function of polarized light scattered from a dielectric film (photoresist) deposited on a glass substrate have been investigated experimentally[7,8]. The vacuum-dielectric interface is a one-dimensional randomly rough interface, while the dielectric-glass interface is planar. A CCD camera was used to record the speckle pattern in the specular direction for each angle of incidence, and the angular intensity correlation function was then calculated from the digitized images. The resulting correlation function displayed two well-defined peaks, one of

which was the memory effect peak, and the other of which was the time-reversed memory effect peak. These peaks contain much the same information as the enhanced backscattering peak. However, their observation is much simpler because they can occur far from the retroreflection direction.

The memory effect has also been observed experimentally in the angular intensity correlation function of light scattered in its double passage through a random phase screen[9,10]. The theory of this effect had been worked out by H. Escamilla, E. R. Méndez, and D. Hotz (Appl. Opt. **32**, 2734 (1993)). The manner in which the speckle pattern is predicted to move as the source is moved is verified experimentally.

D. Angular Correlations in the Intensity of Light Scattered from and Transmitted Through Dielectric and Metal Films with Randomly Rough Surfaces

In recent theoretical work it has been shown that the angular dependence of the intensity of the light scattered from a dielectric film on a reflecting substrate, and in the light scattered from and transmitted through a free-standing metal film, when the illuminated surface in each case is a one-dimensional random surface while the back surface is planar, displays satellite peaks in addition to the enhanced backscattering and enhanced transmission peaks. These satellite peaks occur when the film system supports two or more guided or surface waves at the frequency of the incident light. (A thin metal film supports two and only two surface plasmon polaritons, whose electromagnetic fields are p-polarized.) If the wave numbers of these guided and surface waves are $q_1(\omega), q_2(\omega), \ldots, q_N(\omega)$, the satellite peaks occur at values of the angles of scattering and transmission, θ_s and θ_t, respectively, given by

$$\sin\theta_{s,t} = -\sin\theta_0 \pm \frac{c}{\omega}(q_m(\omega) - q_n(\omega)) \qquad m \neq n, \tag{5}$$

where θ_0 is the angle of incidence. Of course, the satellite peaks will be seen only if the angles θ_s and θ_t defined by Eq. (5) fall in the interval $(-90°, 90°)$, and if their intensity is sufficiently strong. These satellite peaks are multiple-scattering phenomena that arise from the coherent interference of a multiple-scattering sequence in which an intermediate state is the m^{th} guided mode with its time-reversed partner in which an intermediate state is the n^{th} guided wave.

In three recent theoretical studies[11–13] the angular intensity correlations of light reflected from a rough dielectric film on a perfectly conducting substrate,[11,12] or reflected from and transmitted through a rough free-standing metal film[11,13], have been calculated. The resulting correlation functions have a richer structure than the corresponding correlation functions calculated for the light scattered from a semi-infinite metal with a randomly rough surface, which supports only one surface plasmon polariton. In particular, the $C^{(1)}$ and $C^{(2)}$ correlation functions acquire additional peaks whose positions depend on *differences* of the wave numbers of the guided and surface waves. That is, there are now additional memory effect peaks and additional time-reversed memory effect peaks, as well as additional peaks in $C^{(2)}$. Such peaks, of course, have no counterparts in the scattering from the surface of a semi-infinite metal. Additional peaks also occur in the $C^{(1.5)}$ correlation function, but these are due simply to the multiplicity of guided and surface waves in the film structure, and their origin is the same as that of the peaks ini $C^{(1.5)}$ occurring in the scattering from the surface of a semi-infinite metal.

The strengths of these additional peaks are estimated to be large enough for them to be observable. It is hoped that they will be investigated by experimentalists in the near future.

E. Scattering of Electromagnetic Waves from a One-Dimensional Random Metal Surface with a Localized Defect

The scattering of a beam of s-polarized light from a one-dimensional random metal surface with a localized dterministic defect (a narrow Gaussian ridge) is studied by a computer simulation approach[14]. The results show that, for small angles of incidence, the enhanced backscattering peak in the contribution to the mean differential reflection coefficient from the incoherent component of the scattered light is almost unaffected by the presence of the defect. However, for larger angles of incidence the enhanced backscattering peak, is further enhanced by the presence of the defect. This defect-induced enhancement is attributed to a form of the corner cube effect. It is also shown that the memory effect and the time-reversed memory effect peaks in the angular intensity correlation function of the scattered light are enhanced when the light sources and the points of observation are on the same side of the defect, and are suppressed when the light sources and points of observation are on opposite

sides of the defect. The explanation of these effects is again associated with the corner cube effect.

Two experimental investigations have been carried out of the angular intensity correlation function of the light scattered when a polarized beam of light is incident from vacuum on a one-dimensional rough surface[15,16]. One part of the surface used consists of a dielectric film (photoresist) deposited on a glass substrate, while the other part is identical to the first except for the presence of a localized defect on it, a Guassian ridge. The rms height of the random surface was approximately 0.37 μm, and its transverse correlation length was 2.5 μm. The ridge was defined by the profile function $P\exp(-x_1^2/b^2)$, where $P \approx 1.0$ μm and b = 3.2 μm. The correlation function of the intensity of the light scattered from the surface will display a strong correlation when the condition

$$\sin\theta_s - \sin\theta_0 = \sin\theta_s' - \sin\theta_0' \tag{6}$$

is satisfied. This is the condition for the occurrence of the memory and time-reversed memory effects. In the experiment θ_0 was kept fixed and equal to θ_0', while the correlation function was measured as a function of $\Delta\theta_s = \theta_s - \theta_s'$ for a fixed value of θ_s'. The peak in the correlation function at $\Delta\theta_s = 0$ decreased more slowly with increasing $\Delta\theta_s$ for the random surface with a defect than for the random surface without a defect, demonstrating the sensitivity of the angular intensity correlation function to the presence of the defect.

F. Theoretical Studies of Angular Intensity Correlation Functions in Light Scattered from Volume Disordered Dielectric Media

The discovery of the $C^{(10)}$ and $C^{(1.5)}$ correlation functions in the scattering of light from the randomly rough surfaces of semi-infinite metals and of free-standing metal films and dielectric films on reflecting substrates, when the factorization approximation is not made, prompted an examination of whether analogous effects, hitherto unstudied, could be present in the angular intensity correlation functions of light scattered from a volume disordered medium. Such an investigation has been carried out by diagrammatic perturbation theory on the basis of a scalar wave approach[17]. The dielectric constant of the medium is assumed to have the form $\epsilon(\vec{r}) = \epsilon + \delta\epsilon(\vec{r})$, where ϵ is a constant and $\delta\epsilon(\vec{r})$ is a zero-mean, stationary, Gaussian random process defined by $\langle\delta\epsilon(\vec{r})\delta\epsilon(\vec{r}')\rangle = \sigma^2\exp(-|\vec{r}-\vec{r}'|^2/a^2)$, where the angle

brackets denote an average over the ensemble of realizations of $\delta\epsilon(\vec{r})$. The correlation function studied is

$$C(\vec{q},\vec{k}|\vec{q}',\vec{k}') = \langle [I(\vec{q}|\vec{k}) - \langle I(\vec{q}|\vec{k})\rangle][I(\vec{q}'|\vec{k}') - \langle I(\vec{q}'|\vec{k}')\rangle]\rangle \tag{7}$$

where $I(\vec{q}|\vec{k})$ is the intensity of light of wavevector $\vec{k}$ scattered into light of wavevector $\vec{q}$. The $C^{(1)}, C^{(10)}$, and $C^{(1.5)}$ contributions to this correlation function have been calculated. The $C^{(1)}$ contribution, which contains the memory and time-reversed memory effects, has been studied previously on the basis of other models of volume disorder. The $C^{(10)}$ contribution is new. It is proportional to $\delta(\vec{q}-\vec{k}+\vec{q}'-\vec{k}')$, and possesses a structureless dependence on the wave vectors. The contribution $C^{(1.5)}$ is also new, and is unrestricted in its dependence on $\vec{q},\vec{k},\vec{q}',\vec{k}'$. However, it displays several resonance peaks for special linear combinations of $\vec{q},\vec{k},\vec{q}',\vec{k}'$. It is hoped that the results of this study will encourage experimentalists to seek these new features in speckle correlations of light scattered from disordered volume systems.

G. Surface Enhanced Spectral Shifts of Light

It was predicted about a decade ago that the spectrum of the light in the far field emitted by a three-dimensional quasihomogeneous source can differ from that of the source if the spectral coherence of the latter is appropriately chosen (E. Wolf, Nature (London) **326**, 363 (1987); Phys. Rev. Lett. **58**, 2646 (1987); Opt. Commun. **62**, 12 (1987)). Because of the close analogy between the processes of radiation and scattering, this prediction stimulated investigations of spectral changes in the light scattered by volume disordered media. In particular, it was pointed out that the enhanced backscattering of light from a strongly scattering medium, which is due to the coherent interference between multiply-scattered paths and their time-reversed partners, can be regarded as due to the re-emission of light from an extended source in the random medium that possesses just the type of source correlation required to produce a red shift of the spectrum of the scattered light (A. Lagendijk Phys. Lett. A**147**, 389 (1990); Phys. Rev. Lett. **65**, 2082 (1990)). However, the magnitudes of the red shifts calculated for scattering angles close to the backscattering direction were very small, and these shifts remain unobserved experimentally up to the present time.

In order to obtain large enough changes in the spectrum of light scattered from a random medium that they can be measured it is desirable for the angular dependence of the intensity

of the scattered light to possess features that depend strongly on the frequency of the incident light. The backscattering enhancement peak does not possess this feature – it is always in the retroreflection direction – only its width does. A scattering system that does possess such features is a film, either free-standing or supported, that supports two or more guided or surface waves. If the illuminated surface of the film is a one-dimensional randomly rough surface, the angular dependence of the intensity of the light scattered incoherently from the film, or transmitted incoherently through it, will display satellite peaks on both sides of the enhanced backscattering and enhanced transmission peaks (J. A. Sánchez-Gil, A. A. Maradudin, Jun Q. Lu, V. D. Freilikher, M. Pustilnik, and I. Yurkevich, Phys. Rev. **B50**, 15353 (1994); J. Mod. Optics. **43**, 435 (1996); J. A. Sánchez-Gil, A. A. Maradudin, Jun Q. Lu, and V. D. Freilikher, Phys. Rev. **B51**, 17100 (1995)). The scattering angles at which the satellite peaks occur depend on the frequency of the incident light, and this dependence can be quite strong.

In recent theoretical calculations[18] p-polarized light whose spectral density was described by a Gaussian form with a central frequency ω_0 and half-width $\Delta\omega$ was scattered from a ZnS film deposited on the planar surface of a perfect conductor. The illuminated surface of the film was a one-dimensional randomly rough surface. The film supports four guided waves that give rise to eight satellite peaks in the angular dependence of the intensity of the incoherent component of the scattered light, when the light is incident normally on the film, in addition to the enhanced backscattering peak. These satellite peaks give rise to marked shifts away from ω_0 of the position of the maximum of the spectrum of the scattered light for scattering angles in their vicinity. The magnitude of the relative shift in the position $\omega_m(\theta_s)$ of the maximum of the spectrum of the scattered light as a function of the scattering angle θ_s, $|\omega_m(\theta_s) - \omega_0|/\omega_0$, can reach a value as large as 0.012 for θ_s in the vicinity of the angle at which a satellite peak occurs, when $\Delta\omega/\omega_0 = 0.05$. For narrower band widths, corresponding to natural laser sources, the shifts are smaller, e.g. at $\theta_s = 0^\circ (\omega_m - \omega_0)/\omega_0 = -4.4 \times 10^{-4}$ for $\Delta\omega/\omega_0 = 5 \times 10^{-3}$. Even so, they are still about two orders of magnitude larger than those predicted for disordered volume scattering for the same values of $\Delta\omega/\omega_0$.

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